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COOPERATIVE EFFECTS ON TRANSIENT SPECTRAL HOLE BURNING

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Abstract

The nonlinear optical susceptibility is calculated for a system of two closely-located bound excitons by treating the test field as a pertubation in pump-probe experiments. The coherence of the superradiance effect on the spectral hole burning is found to be dramatic in the transient regime for both on-resonance and off-resonance pumping.

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I. Introduction

The pump-probe experiment is one of the most important techniques of nonlinear optics to examine how the optical properties of a medium are modified by a strong laser beam. In the pump-probe process, the strong pump light prepares the material which is subsequently tested by the probe light for information regarding the nonlinear optical response induced by the pump beam. Transient or dynamical spectral hole burning has been observed in such experiments on large molecules in solution, in semiconductor heterostructures and in quasi-one-dimensional conjugated polymers.[1-3]

The phenomenon of hole burning or bleaching accompanied by a dynamical Stark shift in the absorption spectrum has been widely discussed.[4] A phase-space-filling model has been proposed, with a reasonably good explanation provided for the hole burning and excitonic Stark shift observed in semiconductor quantum wells.[4-6] In almost all the existing theoretical treatments[4,7-9] of spectral hole burning, only a single atom is considered in the interacting system.

When the mean distance between the excitons is small compared to the transition wavelength, excitons couple to each other via the exchange of photons. Recently, superradiant effects on the nonlinear optical susceptibility $\chi^{(3)}$ of molecular aggregates in the steady-state regime has been investigated.[10] It is found that there is no universal enhancement of the nonlinearity in general, and that $\chi^{(3)}$ is not enhanced at all under off-resonance conditions.

We investigate, in this article, the cooperative or coherent effect in pump-probe experiments on a semiconductor system of bound-excitons. As a two-atom system is sufficient to demonstrate all the characteristic features of

the coherent effects on the spontaneous radiation,[11] we calculate the pump-field-induced optical response in a system of two closely-located bound excitons with respect to the probe-field in the transient regime.

Since the probe field is much weaker than the pump field in typical pump-probe experiments, we solve the wave equation to all orders of the pump field and first order of the probe field in our calculation of the susceptibility experienced by the probe field. Coherence effects are obtained by comparing our results with those from the corresponding incoherent excitons. In contrast to the conclusion of Ref. 10, we find a very different nonlinear optical susceptibility. Its decay rate and Rabi oscillation frequency, as well as the phenomenon of spectral hole burning, all show dramatic changes due to the coherent effect in the two-coupled excitons.

II. Method of Calculation

The nonlinear optical response of a solid can be treated by the two-level model. The two levels in a semiconductor correspond to the ground state $|->$ (electron-hole recombination) and the bound exciton state $|+>$. Since bound excitons trapped by impurities cannot propagate, the two-exciton system may be described as two stationary atoms. A strong laser pump field and a much weaker laser probe field are directed to the semiconductor. As long as the laser wavelengths are much larger than the separation between the excitons, it is a good approximation to assume the same field is experienced by both excitons. Thus, the Hamiltonian can be written, in the rotating-wave approximation, as

$$H = \omega_x \sum_{j=1}^{2} S_j^z - [\mu(E_p^* e^{i\omega_p t} + E_t^* e^{i\omega_t t}) \sum_{j=1}^{2} S_j + H.c.] \quad , \tag{1}$$

where we have defined the dipole operator

$$S_j = |->_j \, {}_j\!<+| \quad , \tag{2a}$$

$$S_j^z = \frac{1}{2}(|+>_j \, {}_j\!<+| - |->_j \, {}_j\!<-|) \quad . \tag{2b}$$

The matrix element of the excitonic dipole moment is μ, the exciton frequency is ω_x, and E_p (E_t) is the amplitude of the pump (probe) field with frequency ω_p (ω_t).

Let us now define the four states for the two-exciton system,

$$|1> = |-,-> \quad , \quad |2> = |+,-> \quad , \quad |3> = |-,+> \quad , \quad |4> = |+,+> \quad . \tag{3}$$

At an arbitrary time t, the state for the system can be written as

$$|\psi(t)> = C_1(t)|1> + C_2(t)|2> + C_3(t)|3> = C_4(t)|4> \quad , \tag{4}$$

where the coefficients are determined by the set of coupled equations

$$\dot{C}_1 = i\omega_x C_1 + i(\Omega_p e^{i\omega_p t} + \Omega_t e^{i\omega_t t})(C_2 + C_3) \quad , \tag{5a}$$

$$\dot{C}_2 = i(\Omega_p e^{-i\omega_p t} + \Omega_t e^{-i\omega_t t})C_1 - \gamma C_2 + i(\Omega_p e^{i\omega_p t} + \Omega_t e^{i\omega_t t})C_4 \quad , \tag{5b}$$

$$\dot{C}_3 = i(\Omega_p e^{-i\omega_p t} + \Omega_t e^{-i\omega_t t})C_1 - \gamma C_3 + i(\Omega_p e^{i\omega_p t} + \Omega_t e^{i\omega_t t})C_4 \quad , \tag{5c}$$

$$\dot{C}_4 = i(\Omega_p e^{-i\omega_p t} + \Omega_t e^{-i\omega_t t})(C_2 + C_3) - (i\omega_x + 2\gamma)C_4 \quad . \tag{5d}$$

In Eqs. (5), we have assumed that μ and $E_{p,t}$ are all real quantities, and have defined the Rabi frequency $\Omega_{p,t} = \mu E_{p,t}$. We have also introduced phenomenologically the nonradiative decay rate γ which is a result of the strong Auger effect for bound excitons in semiconductors.[13] For convenience, we work in the rotating frame by making the replacements $C_1 \rightarrow C_1 e^{i\omega_p t}$ and $C_4 \rightarrow C_4 e^{-i\omega_p t}$, whereby Eqs. (5) become

$$\dot{C}_1 = -i\Delta C_1 + i(\Omega_p + \Omega_t e^{-i\Delta_t t})(C_2 + C_3) \quad , \tag{6a}$$

$$\dot{C}_2 = i(\Omega_p + \Omega_t e^{i\Delta_t t})C_1 - \gamma C_2 + i(\Omega_p + \Omega_t e^{-i\Delta_t t})C_4 \quad , \tag{6b}$$

$$\dot{C}_3 = i(\Omega_p + \Omega_t e^{i\Delta_t t})C_1 - \gamma C_3 + i(\Omega_p + \Omega_t e^{-i\Delta_t t})C_4 \quad , \tag{6c}$$

$$\dot{C}_4 = i(\Omega_p + \Omega_t e^{i\Delta_t t})(C_2 + C_3) + (i\Delta - 2\gamma)C_4 \quad . \tag{6d}$$

with the detuning parameters defined by $\Delta = \omega_p - \omega_x$, and $\Delta_t = \omega_p - \omega_t$.

To solve Eqs. (6), we treat the probe field as a perturbation and obtain the zeroth-order solutions by setting $\Omega_t = 0$. The results are

$$C_1^{(0)}(t) = e^{-\gamma t}\{\frac{(\Delta+i\gamma)^2+2\Omega_p^2}{\nu^2}\cos\nu t - i\frac{\Delta+i\gamma}{\nu}\sin\nu t + \frac{2\Omega_p^2}{\nu^2}\} \quad , \tag{7a}$$

$$C_2^{(0)}(t) = C_3^{(0)}(t) = \frac{\Omega_p}{\nu^2} e^{-\gamma t}[(\Delta+i\gamma)(1-\cos\nu t) + i\nu\sin\nu t] \quad , \tag{7b}$$

$$C_4^{(0)}(t) = \frac{2\Omega_p^2}{\nu^2} e^{-\gamma t}(1-\cos\nu t) \quad . \tag{7c}$$

with the initial condition $C_1^{(0)}(0) = 1$. If the system is initially in the state $|2\rangle$, the zeroth-order solutions are given by

$$C_1^{(0)}(t) = \frac{\Omega_p}{\nu^2} e^{-\gamma t}[i\nu\sin\nu t + (\Delta+i\gamma)(1-\cos\nu t)] \quad , \tag{8a}$$

$$C_2^{(0)}(t) = e^{-\gamma t}[1-\frac{2\Omega_p^2}{\nu^2}(1-\cos\nu t)] \quad , \tag{8b}$$

$$C_3^{(0)}(t) = -\frac{2\Omega_p^2}{\nu^2} e^{-\gamma t}(1-\cos\nu t) \quad , \tag{8c}$$

$$C_4^{(0)}(t) = \frac{\Omega_p}{\nu^2} e^{-\gamma t}[i\nu\sin\nu t - (\Delta+i\gamma)(1-\cos\nu t)] \quad . \tag{8d}$$

In these equations, we have defined a new complex Rabi frequency

$$\nu = [(\Delta+i\gamma)^2 + 4\Omega_p^2]^{\frac{1}{2}} \quad . \tag{9}$$

Following the procedure discussed in Ref. 14, we find the solutions up to the first order in Ω_t by substituting Eq. (7) or (8) into (6), depending on the initial condition considered. The results are given by

$$C_i(t) = C_i^{(0)}(t) + C_i^{(1)}(t) \quad , \qquad i = 1,2,3,4 \quad . \tag{10}$$

For the initial condition $C_1(0) = 1$, we have

$$C_1^{(1)}(t) = \frac{\Omega_t\Omega_p}{\nu^2} e^{-i(\Delta_t - i\gamma)t} \times \left\{\frac{[\nu^2-(\Delta+i\gamma)(\Delta_t-i\gamma)]\cos\nu t+i\nu(\Delta_t-\Delta-2i\gamma)\sin\nu t}{\nu^2-(\Delta_t-i\gamma)^2} - \frac{\Delta+i\gamma}{\Delta_t-i\gamma}\right\} \quad , \tag{11a}$$

$$C_2^{(1)}(t) = \frac{\Omega_t}{\nu^2} e^{(i\Delta_t-\gamma)t}\left\{\frac{2\Omega_p^2}{\Delta_t+i\gamma} + \frac{i\nu[(\Delta+i\gamma)^2+2\Omega_p^2+(\Delta+i\gamma)(\Delta_t+i\gamma)]\sin\nu t-\{\nu^2(\Delta+i\gamma)+(\Delta_t+i\gamma)[(\Delta+i\gamma)^2+2\Omega_p^2]\}\cos\nu t}{\nu^2-(\Delta_t+i\gamma)^2}\right\}$$

$$+ \frac{2\Omega_t\Omega_p^2}{\nu^2} e^{-(i\Delta_t+\gamma)t}\left[\frac{1}{\Delta_t-i\gamma} + \frac{(\Delta_t-i\gamma)\cos\nu t+i\nu\sin\nu t}{\nu^2-(\Delta_t-i\gamma)^2}\right] = C_3^{(1)}(t) \quad , \tag{11b}$$

$$C_4^{(1)} = \frac{2\Omega_t\Omega_p}{\nu^2} e^{(i\Delta_t-\gamma)t}\left[\frac{\Delta+i\gamma}{\Delta_t+i\gamma} + \frac{[(\Delta+i\gamma)(\Delta_t+i\gamma)+\nu^2]\cos\nu t-i\nu(\Delta+\Delta_t+2i\nu)\sin\nu t}{\nu^2-(\Delta_t+i\gamma)^2}\right] . \tag{11c}$$

When the initial condition is $C_2(0) = 1$, we have

$$C_1^{(1)}(t) = \frac{\Omega_t}{\nu^2} e^{-(i\Delta_t+\gamma)t}\left\{\frac{4\Omega_p^2}{\nu^2-(\Delta_t-i\gamma)^2}[(\Delta_t-i\gamma)\cos\nu t+i\nu\sin\nu t]-\frac{(\Delta+i\gamma)^2}{\Delta_t-i\gamma}\right\} \quad , \tag{12a}$$

$$C_2^{(1)}(t) = \frac{\Omega_p\Omega_t}{\nu^2} e^{(i\Delta_t-\gamma)t}\left\{\frac{\Delta+i\gamma}{\Delta_t+i\gamma} + \frac{[\nu^2+(\Delta+i\gamma)(\Delta_t+i\gamma)]\cos\nu t-i\nu(\Delta+\Delta_t+2i\gamma)\sin\nu t}{\nu^2-(\Delta_t+i\gamma)^2}\right\}$$

$$+\frac{\Omega_p\Omega_t}{\nu^2}e^{-(i\Delta_t+\gamma)t}\{\frac{\Delta+i\gamma}{\Delta_t-i\gamma}+\frac{[\nu^2+(\Delta+i\gamma)(\Delta_t-i\gamma)]\cos\nu t+i\nu(\Delta+\Delta_t)\sin\nu t}{\nu^2-(\Delta_t-i\gamma)^2}\}$$

$$=C_3^{(1)}(t) \quad , \tag{12b}$$

$$C_4^{(1)}(t)=\frac{\Omega_t}{\nu^2}e^{(i\Delta_t-\gamma)t}[\frac{(\Delta+i\gamma)^2}{\Delta_t+i\gamma}+4\Omega_p^2\frac{i\nu\sin\nu t-(\Delta_t+i\gamma)\cos\nu t}{\nu^2-(\Delta_t+i\gamma)^2}] \quad . \tag{12c}$$

We note that in the integration of Eqs. (8), the static terms have been neglected, and as a consequence, we have $C_2^{(1)}(t)=C_3^{(1)}(t)$.

The expectation values of the dipole moment of the excitons are

$$d_1=\mu\langle S_1(t)\rangle=\mu(C_1^*C_2+C_3^*C_4) \quad , \tag{13a}$$

$$d_2=\mu\langle S_2(t)\rangle=\mu(C_1^*C_3+C_2^*C_4) \quad . \tag{13b}$$

The dipole moments induced by the probe laser field are therefore

$$\delta d_1=\mu(C_1^{(0)*}C_2^{(1)}+C_2^{(0)}C_1^{(1)*}+C_3^{(0)*}C_4^{(1)}+C_4^{(0)}C_3^{(1)*}) \quad , \tag{14a}$$

$$\delta d_2=\mu(C_1^{(0)*}C_3^{(1)}+C_3^{(0)}C_1^{(1)*}+C_2^{(0)*}C_4^{(1)}+C_4^{(0)}C_2^{(1)*}) \quad . \tag{14b}$$

The nonlinear optical susceptibility of the two-exciton system as experienced by the probe field is by definition

$$\chi_t=\mu\sum_{j=1}^{2}n_j\delta d_j/E_te^{-i\omega_t t} \quad , \tag{15}$$

where

$$n_1 = |C_2|^2 + |C_4|^2 \approx |C_2^{(0)}|^2 + |C_4^{(0)}|^2 \tag{16a}$$

$$n_2 = |C_3|^2 + |C_4|^2 \approx |C_3^{(0)}|^2 + |C_4^{(0)}|^2 \tag{16b}$$

are the exciton population probabilities.

Since we are only interested in solutions up to first order in E_t, the terms leading to the static and second-harmonic components in Eqs. (14) can be neglected. The coherent effect on the susceptibility due to the cooperative coupling between the two excitons can be most easily found by comparing the above results with those of two independent excitons. The corresponding susceptibility for two independent or incoherent excitons is

$$\chi_t^i = \frac{16\Omega_p^2\mu^2}{|\nu|^2} e^{-2\nu t} \left|\sin\frac{\nu}{2}t\right|^2 \left([\nu^* \cos\frac{\nu^*}{2}t + i(\Delta - i\gamma)\sin\frac{\nu^*}{2}t] \right.$$
$$\frac{i[\nu^2 + (\Delta+i\gamma)(2\Delta_t - \Delta + i\gamma)]\sin\frac{\nu}{2}t - 2\gamma(\Delta_t + i\gamma)\cos\frac{\nu}{2}t}{\nu^2 - (2\Delta_t - \Delta + i\gamma)^2}$$
$$\left. - 4\Omega_p^2 \sin\frac{\nu}{2}t \frac{i\nu^* \cos\frac{\nu^*}{2}t + (2\Delta_t + \Delta + i\gamma)\sin\frac{\nu^*}{2}t}{\nu^{*2} - (2\Delta_t + \Delta - i\gamma)^2} \right) . \tag{17}$$

where we have used the initial conditions that there is no exciton initially.

III. Results and Discussion

As can be seen from Eqs. (7), (8), (11), (12) and (17), there exist optical nutations in the susceptibility χ_t. In the coupled two-exciton case, there are four oscillations with frequencies $Re\nu$, $2Re\nu$, $3Re\nu$ and $4Re\nu$

composing the Rabi oscillation, while in the incoherent case the oscillation consists of only two components with frequencies $Re\nu$ and $2Re\nu$.

When $\Omega_p \leq \gamma/2$, or when the non-radiative decay is faster than the flip-flop rate, χ_t decays monotonically, and its rate is generally faster than the decay of χ_t^i. As an example, one can easily see that for $\Delta = 0$ and $\Omega_p = \gamma/2$ the decay factor is $e^{-2\gamma t}$ for χ_t^i, but is $e^{-4\gamma t}$ for χ_t because of the superradiant effect in the coupled system.

The imaginary part of the optical susceptibility as given by Eqs. (15) and (17) is computed numerically under different conditions. We adopt the unit such that energy is measured by γ and time is measured by γ^{-1} throughout this paper. When the pump field is tuned on resonance with the exciton frequency, or when $\Delta = 0$, the cooperative effect is in general found to make the hole shallow and at the same time to make χ_t decay faster. This is clearly seen in Fig. 1 in which $Im\chi_t(t)$ is plotted for the coupled two-exciton system with $C_1(0) = 1$ (a) and $C_2(0) = 1$ (b) as well as for incoherent excitons (c). Holes induced by the pump-field bleaching are located at $\omega_t = \omega_x$. We have also found the phenomenon of power broadening in spectral hole burning in our numerical study, that is, a stronger pump intensity leads to broader holes. It is further observed from Fig. 1(b) that when one of the excitons is already excited initially, two antiholes appear on both sides of the hole. This means that bleaching or increasing in absorption depends on the initial conditions of the system.

For the pump field tuned off-resonance with the exciton frequency, i.e., $\Delta \neq 0$, we plot the absorption spectra in Fig. 2 for $C_1(0) = 1$, in Fig. 3 for $C_2(0) = 1$, and in Fig. 4 for incoherent case. A comparison of 2(b) with 4(b) reveals that the superradiance effect can enhance the spectral hole burning which is centered around $\omega_t = \omega_x$. The situation becomes much more complicated

when there is initially one exciton already excited. In addition to holes at $\omega_t = \omega_x$, there are also holes and antiholes appearing near $\omega_t = 2\omega_p - \omega_x$, as shown in Fig. 3. This is a result of a two-photon effect of the pump-field. In a study of Frankel excitons in Ref. 10, no enhancement in the nonlinear optical response was found for the case of off-resonance pumping. On the contrary, for bound excitons we find an important qualitative difference in the optical hole burning or bleaching for the off-resonance pumping, as well as the hole burning and broadening for the on-resonance pumping.

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Figure Captions

1. Evolution of the absorption spectrum of the susceptibility of two excitons experienced by the probe field for resonant pumping ($\Delta = 0$) and $\Omega_p = 0.4$, where Ω_p and Δ_t are in the unit of γ, t in γ^{-1}, and χ_t and χ_t^i in μ^2. (a) $Im\chi_t$ with the initial condition $C_1(0) = 1$; (b) $Im\chi_t$ with $C_2(0) = 1$; (c) $Im\chi_t^i$ with the initial conditions of no excitons.
2. Evolution of the absorptive spectrum of the susceptibility $Im\chi_t$ with off-resonant pumping ($\Delta = -5$) and $C_1(0) = 1$, where Δ, Δ_t and Ω_p are in the unit of γ, t in γ^{-1}, and χ_t in μ^2. (a) $\Omega_p = 0.4$; (b) $\Omega_p = 1$.
3. Same as Fig. 2, except that $C_2(0) = 1$.
4. Same as Fig. 2, except that χ_t is replaced by χ_t^i.

Fig. 1

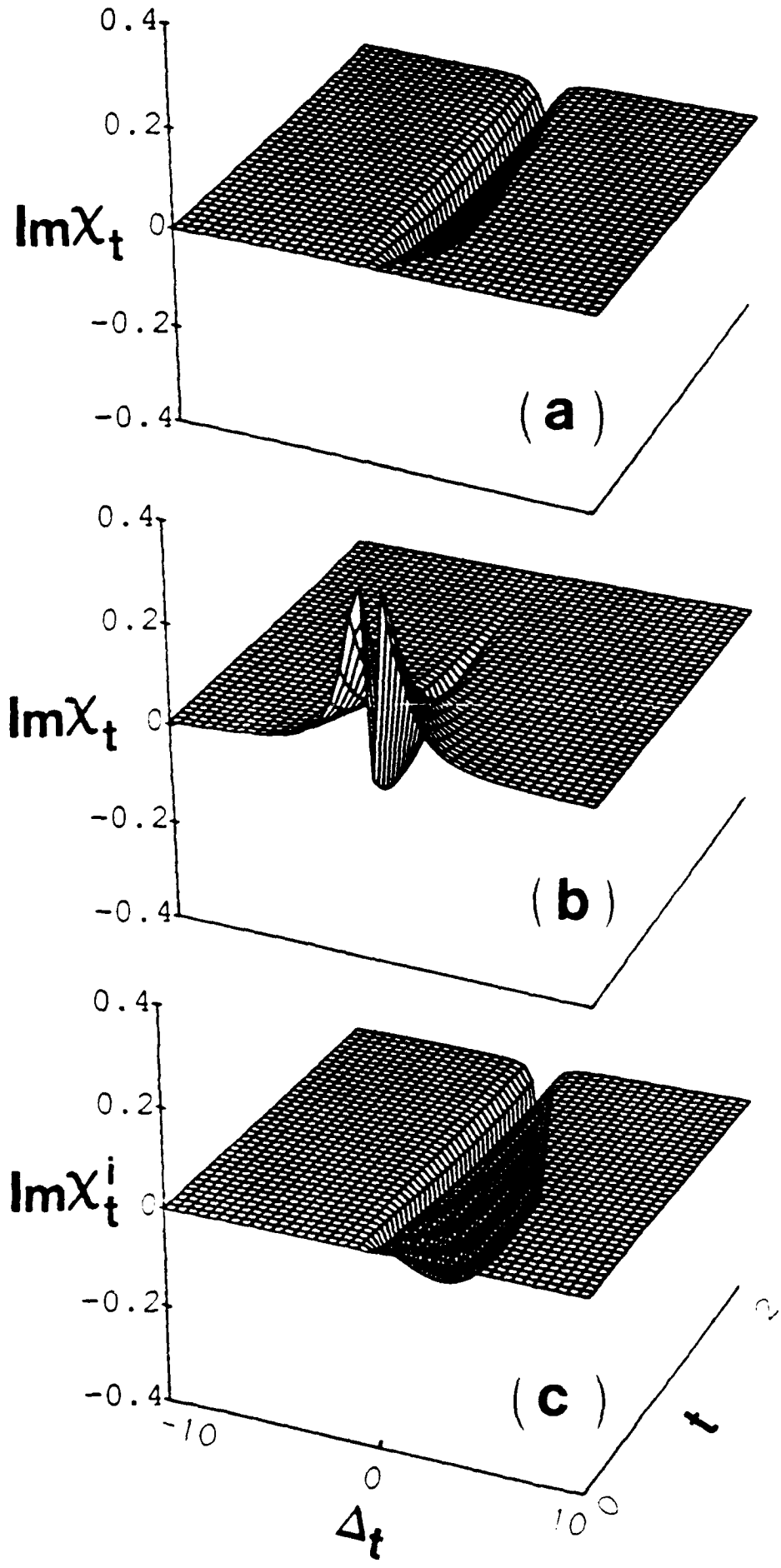

Fig. 2

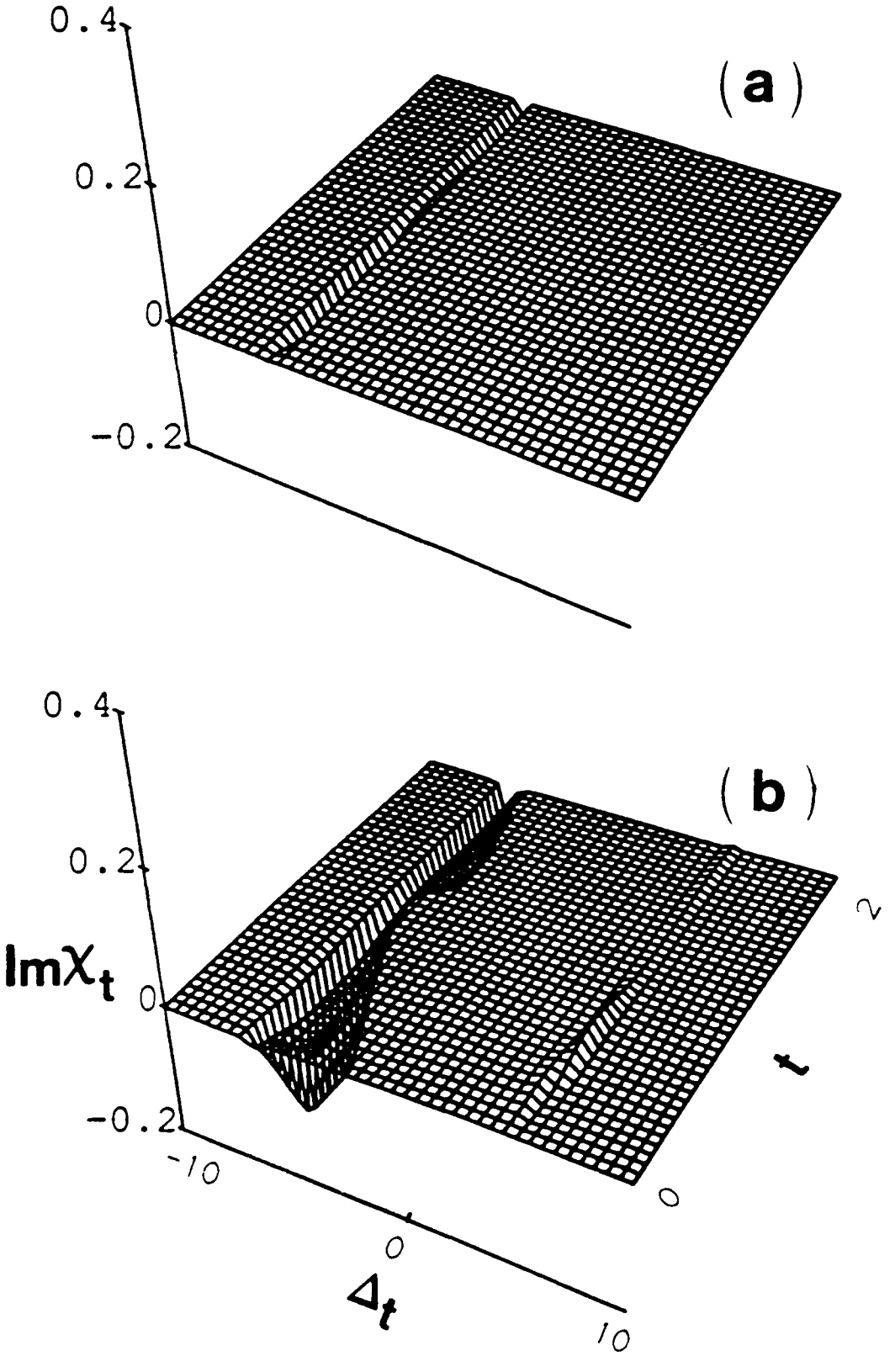

Fig. 3

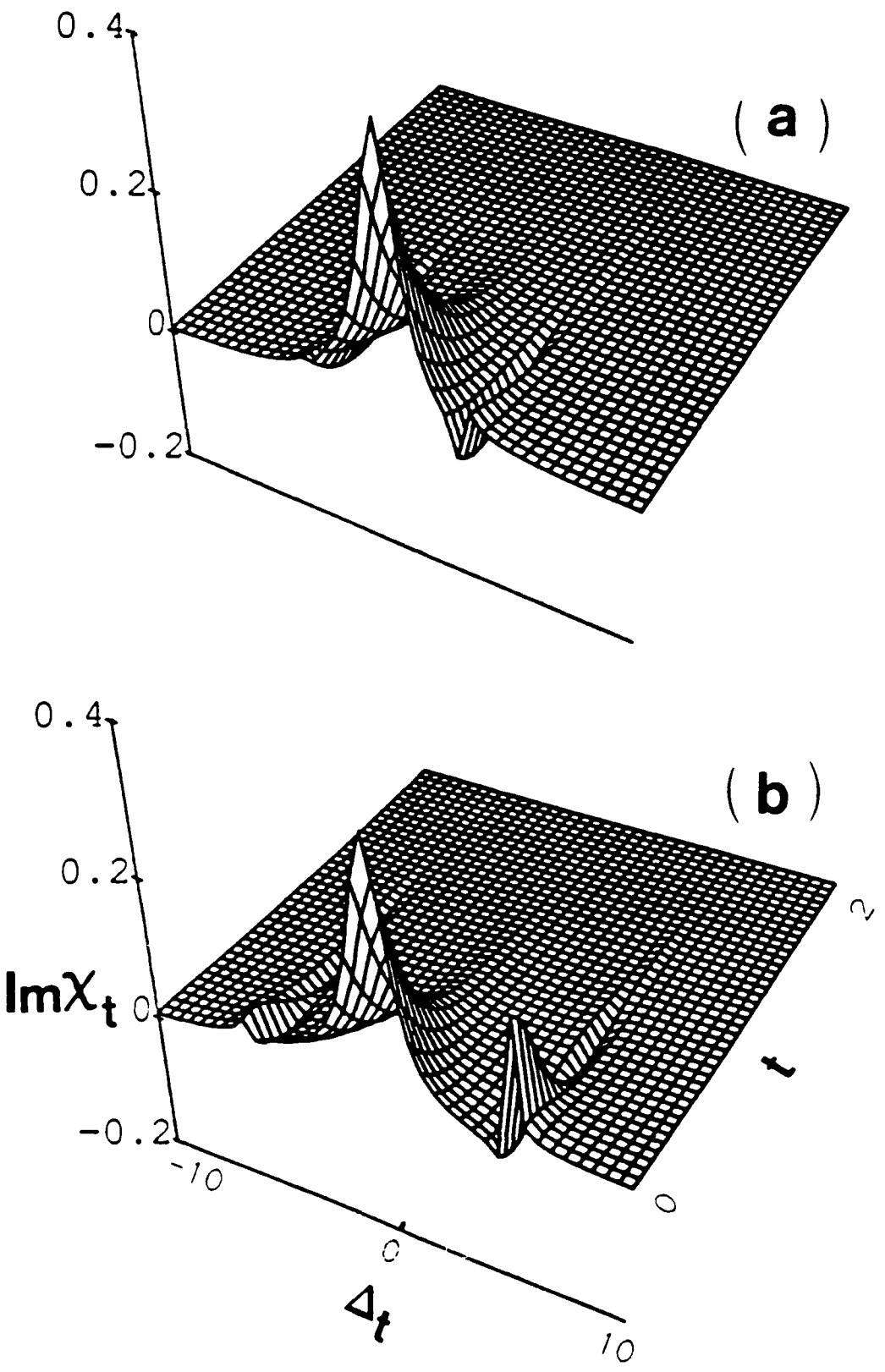

Fig. 4